Photoconduction and photocontrolled collective effects in the Peierls conductor TaS_3

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Light illumination of thin crystals of CDW conductor TaS_3 is found to result in dramatic changes of both linear (G) and nonlinear conduction. The increase of G is accompanied by suppression of the collective conduction, growth of the threshold field E_T, and appearance of the switching and hysteretic behavior in the nonlinear conduction. The effects in the nonlinear conduction are associated with increase of CDW elasticity due to illumination that leads in particular to appearance of a relation $E_T \propto G^{1/3}$ expected for the one-dimensional pinning.

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Quasi-one-dimensional (quasi-1D) conductors with charge-density-waves (CDW) [1] are one of the most interesting physical systems with collective electron transport. The interaction between electrons condensed into the CDW dominates in elastic properties of the electron crystal — CDW. The elastic properties of the CDW affect such characteristics of quasi-1D conductors as the value of the threshold field for CDW sliding, E_T, phase-correlation length, dielectric constant, *et al.* In its turn the elastic properties are controlled by quasiparticles (electrons and holes) thermally excited over the Peierls gap in the energy spectrum and screening the electric fields caused by CDW deformations. Thus a variation of the quasiparticle concentration (or the total carrier concentration) may be a tool controlling the properties of CDW conductors. An attempt to vary the total carrier concentration has been undertaken in the field effect experiment [2]. In particular it was found that 1%-change of the total concentration of the current carriers by the transverse electric field leads to 40%-change of the threshold field value. Another well-known way to modify the carrier concentration is excitation of nonequilibrium current carriers by light. For example, illumination of a semiconductor may result in increase of the carrier concentration by orders of magnitude. Such a change can be easily detected as a variation of the conduction (photoconduction). Photoconduction is one of the most fruitful methods to study the details of the energy structure, current carrier recombination time, and other semiconductor parameters. The similarity of numerous physical properties of the CDW conductors and semiconductors arising from the existence of the gap in the electron state density is well known [3].

Several attempts of experimental search for photoconduction of CDW materials [4, 5, 6, 7] reveal contradictory results. In Ref. [4, 5] no noticeable photoconduction in TaS_3 was observed. Instead, the bolometric response was found and employed for detailed study of the energy structure in TaS_3. In addition an enhancement of the bolometric response was reported in nonlinear regime Ref. [4]. In Ref. [6] photoinduced CDW conduction was observed in blue bronze $K_{0.3}MoO_3$. The red boundary of the effect was found to correspond to the Peierls gap value. The phenomenon was associated with initiation of the CDW depinning by optically excited single electrons. No light-induced variation of the linear conduction and the threshold field was reported. In Ref. [7] photoinduced modification of the dynamic transition from slide to creep in $K_{0.3}MoO_3$ was reported: light illumination was found to increase E_T and the CDW creep rate. The origin of the effect was attributed to a local destruction of the CDW which led to the photoinduced phase slip and the redistribution of the CDW phase. No effect of illumination on the linear conduction was reported. Thus, despite some similarity between CDW conductors and semiconductors no photoresponse in the linear conduction was found during 25 years of study of the CDW materials. Its worth to mention that the absence of photoconduction would agree with theories predicting very small quasiparticle lifetime, of the order of 10^{-12} s [8] Femtosecond spectroscopy study of $K_{0.3}MoO_3$ has shown that the electron-hole recombination time is short indeed, of subpicosecond scale [9]. From this point of view quasiparticles (electrons and holes) are ill-defined physical objects and the absence of photoconduction is a feature of CDW conductors. So observation of photoconduction is essential for physics of CDW conductors.

Here we show that photoconduction of quasi-1D conductor TaS_3 can be directly observed in low-frequency conduction measurements. In particular, we show that the linear conduction may be increased up to an order of magnitude under the light illumination. The resulting changes of the nonlinear conduction are dramatic, and reveal themselves in a substantial growth of the threshold field, suppression of the nonlinear conduction near E_T, and appearance of the switching behavior. We also show that the observed growth of E_T can entirely accounted by photoinduced increase of the CDW elasticity. Thus, no exotic assumptions on photoinduced phase slip [7] or CDW depinning [6] is required. Moreover, our results clearly demonstrate the opposite effect, suppression of the collective conduction under light illumination.

Orthorhombic TaS_3 is a typical Peierls conductor. In this material the CDW formation at $T_P = 220$ K is accompanied by the complete dielectrization of the electron spectrum. All five studied samples (made by split-

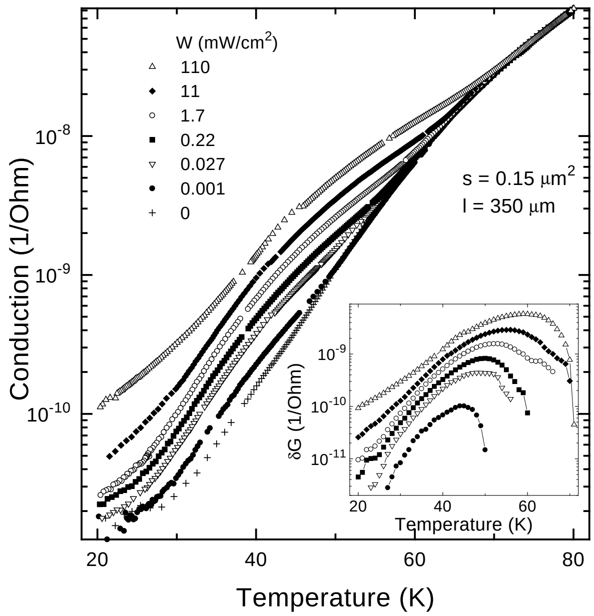

FIG. 1: Temperature dependencies of conduction, G, at different steady light illumination intensities, W. The inset shows respective temperature dependencies of photoconduction, $\delta G = G(T, W) - G(T, 0)$. All the curves were obtained upon cooling to eliminate the conduction hysteresis.

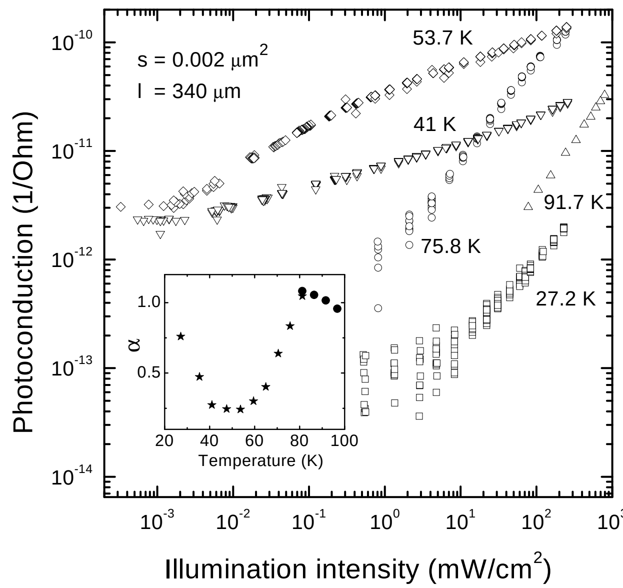

FIG. 2: The dependences of photoconduction δG on the light illumination intensity W at different temperatures. The inset shows the temperature dependence of the exponent $\alpha = d \ln \delta I / d \ln W$ (different marks correspond to the different methods of the measurements, see the text for detailes).

ting of high-quality crystals) having cross-section areas $0.002 \leq s \leq 0.15$ μm^2 demonstrated qualitatively similar behavior. Such thin crystals were chosen to enhance the contribution of the region, affected by the illumination [10]. In our samples the photoconduction developed in the entire sample, rather than in a surface layer shunted by bulk like in Refs. [4, 5, 6, 7]. Using of thin samples allows also to suppress heating effect due to exceptionally good thermal contact with sapphire substrate. In addition, growth of potential relief due to finite-size effects [13] leads to substantial photocondution growth due to spatial separation of photoexcited electrons and holes. Current terminals were made by indium cold soldering, all measurements were performed in the two-terminal regime. IR LED with intensities (10^{-6} - 1) W/cm^2 at the sample position, and with a wavelength of $\lambda = 0.94$ μm was used. Thus the photon energy was higher than the optical gap value 125 meV [5].

Fig. 1 shows a set of temperature dependencies of conduction, $G(T, W)$, at various intensities W of steady light illumination for the most thick sample. Noticeable deviation from the darkness curve starts at $T < 70$ K. The inset shows the respective set the photoconduction, $\delta G = G(T, W) - G(T, 0)$. The position of maximum of $\delta G(T)$ varies with W in the range 40 - 65 K. The deviation starts at somewhat higher temperatures for thinner samples. For example, for the thinnest one having $s = 0.002$ μm^2 $\delta G/G \sim 0.01\%$ was observed at $T = 100$ K. Maximum of $\delta G(T)$ at $W = 110$ mW/cm^2 is at $T = 65$ K. The upper boundary of the light-induced heating may be estimated using $\delta G(T)$ at $T = 100$ K to be as small as $(\delta G/G)/(dG/dT) \sim 1$ mK, as a consequence of exceptionally good thermal contact between the sample and substrate. All the results presented below were obtained for this thinnest sample having the length $l = 340$ μm and the room-temperature resistance $R_{300} = 430$ kOh.

The dependence of the value of photoconduction δG on the light illumination intensity W was studied by two methods. At the temperature range 27 - 95 K AC conduction was measured at the frequency $f = 4.5$ Hz as a function of intensity W of a steady light illumination. Such a low frequency value was chosen to enable measurements of very low conduction values. At temperatures $T > 90$ K the ratio $\delta G/G$ becomes too small and comparable with one resulting from temperature fluctuations. To improve measurements quality the double modulation method was used at $T = 81$ - 100 K. Namely, the modulation of AC conduction due to the light illumination was detected at the frequency of light chopping $f_{ch} = 4.5$ Hz, AC conduction being measured at the frequency $f = 335$ Hz. The results are presented in Fig. 2. The dependencies can be approximated by the power law $\delta I = W^\alpha$, where the photocurrent $\delta I = \delta G V$. These dependencies are highly nonlinear in the middle of the temperature range ($\alpha < 1$), and approach to the linear ones ($\alpha \approx 1$) at its ends. The temperature dependence of the exponent α determined as $\alpha = d \ln \delta I / d \ln W$ at $W > 10$ mW/cm^2 is shown in the inset in Fig. 2. This

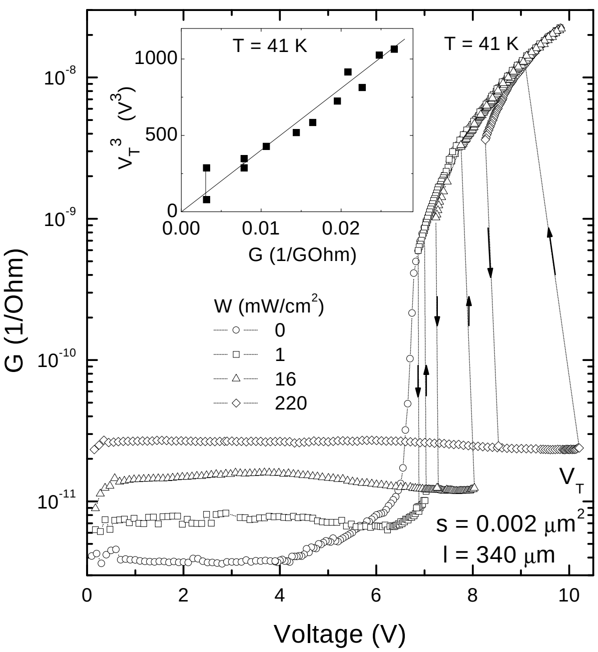

FIG. 3: The dependencies of conduction $G = I/V$ on the sample voltage V at different steady illumination intensities W. The arrows show directions of the current sweep.The inset shows the dependence V_T^3 on linear conduction G (at 100 mV) at different light intensities. Vertical lines correspond to uncertaincy in determination of V_T at $W = 0$ and $W = 1$ mW/cm^2.

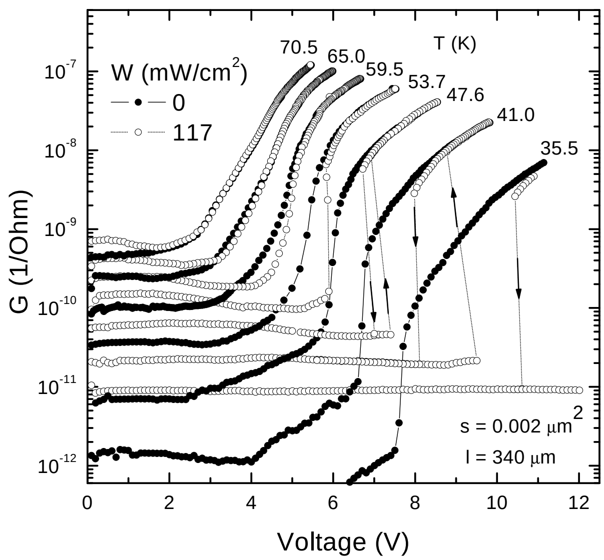

FIG. 4: The dependencies of conduction G on the sample voltage V at different temperatures. The dark circles correspond to the measurements in the darkness, the open circles correspond to the ones under steady light illumination.

dependence has a pronounced minimum at $T = 50$ K where the change of the light intensity up to 4 orders of magnitude leads to the increase of the linear conduction only up to an order of magnitude.

At sufficiently low temperatures the light-induced conduction variation becomes so large that it can be clearly seen in I-V curves. Fig. 3 shows the evolution of I-V curves (plotted as $G = I/V$ *vs.* V) caused by light illumination at $T = 41$ K. In the darkness the $G(V)$ curve has the usual shape: there is a region of a constant (linear) conductivity in a small electric field, then a region of creep (weak nonlinearity), and a region of CDW sliding (strong nonlinearity). The curve has a smooth character without any switching and hysteresis. The dramatic changes of the shape of $G(V)$ curves both in the linear and nonlinear conduction take place under light illumination. It can be seen that the growth of the light intensity causes
1) increase of the linear conduction up to 10 times,
2) decrease of the conduction with voltage growth in the creeping region,
3) increase of up to 60% in the threshold field for the onset of CDW sliding
(decrease of the nonlinear conduction near V_T), and
4) appearance of the switching regime (unusual for TaS_3 samples) with hysteretic character [11]. The growth of the threshold field and appearance of the switching under light illumination was also observed in $K_{0.3}MoO_3$ [7].

Fig. 4 shows the temperature evolution of $G(V)$ dependencies measured under steady light illumination ($W = 117$ mW/cm^2) and in the darkness. At temperatures above 65 K (corresponding to the maximum of the value of photoconduction for this sample) the changes of the shape of $G(V)$ curves under light illumination are visible only in the linear conduction region and in the region of creep. With temperature decrease the dependencies $G(V)$ under light illumination begin to deviate from the dark ones in the CDW sliding region as well. The transition to the sliding regime becomes more sharp, the switching behavior appears, and the dramatic growth of the width of the hysteresis loop develops.

It is worth to note that all the $G(V)$ curves coincide in the high voltage region $E \gg E_T$. This indicates that 1) the relative variation of the nonlinear conduction due to illumination is much less than for the linear conduction, 2) the heating of the sample due to illumination is small indeed (heating leads to an increase of the conduction and decrease of the threshold field).

Our results allow to estimate the recombination time τ of photoexcited carriers in TaS_3. The photoexcited current carrier concentration Δn is determined by the balance between the rate of their photogeneration, $kaLn_{ph}(1 - e^{-\beta b})$, and the relaxation rate $abL\Delta n/\tau$ (we assume $\Delta n \ll n$), where k is the quantum efficiency of photogeneration, n_{ph} is the number of incident photons per area per unit time, β is the absorption coefficient, and a, b and L are the sample width, thickness

and length, respectively. Thus, for $\beta b \ll 1$ one gets $\tau = \Delta n / k\beta n_{ph}$. At $T = 100$ K $\Delta n (\propto \delta G) \propto n_{ph} (\propto W)$, i.e. τ is independent of the light intensity W. As mentioned above, for this sample $\delta G/G \sim 0.01\%$ at $T = 100$ K. As $\Delta n/n(T) = \delta G/G(T)$, $n_{100} \approx 10^{-3} n_{300}$, $n_{300} \approx 2 \times 10^{21}$ cm^{-3}, so $\Delta n \approx 2 \times 10^{14}$ cm^{-3}. The value of k is unknown. Assuming $k = 1$, $1/\beta = 0.3$ μm [10], for $W = 110$ mW/cm^2 one gets $\tau_{100} \approx 10^{-10}$ s. Moreover, this value is almost two orders of magnitude greater at 65 K and $W = 110$ mW/cm^2 (see Fig. 1). This time is much smaller than that in pure semiconductors. This explains why the photoconduction in quasi-1D conductors was not found during a long time. On the other hand, τ in thin crystals of TaS_3 is orders of magnitude greater $\tau \approx 5 \times 10^{-13}$ s measured by optical methods in blue bronze at the same temperatures [9]. Possible physical mechanism providing enhancement of photoconduction in thin samples is given in the following paragraph.

In the temperature range 30 K$< T <$ 70 K there is a nonlinear relation between the light intensity and photoconduction (see inset in Fig. 2), and the photoconduction reaches its maximum (inset in Fig. 1). These means that in this temperature range the recombination time of photoexcited current carriers 1) depends on their concentration, 2) is larger than that out of this temperature range. These features are peculiar to so-called "persistent photoconduction" (also "delayed" or "frozen") well-known for some inhomogeneous semiconductors [12]. In our case the barriers for recombination result from the potential relief caused by the pinned CDW. The energy range for this relief can be estimated from the relation $\delta\zeta \sim E_T L_\parallel$, where $L_\parallel$ is CDW phase correlation length. At higher temperatures, E_T is getting smaller (see Fig. 4), and finally one gets $\delta\zeta \ll T$. As a result, the relaxation barriers diminish and the relaxation is getting faster with a rate practically independent of the light intensity. As the threshold field rapidly increases with temperature decrease, so a situation when $\delta\zeta$ is of the order of the Peierls gap is achieved. In this case one may expect nucleation of dislocations and opening of a new relaxation channel. This may explain the low-temperature decrease of photoconduction (see inset in Fig. 1). In addition, since E_T is illumination-dependent, the barrier height depends on the concentration of photoexcited current carriers. Taking $L_\parallel = 1$ - 10 μm [13] and $E_T \approx 30$ mV/μm for $T = 41$ K at $W = 220$ mW/cm^2 (see Fig. 3) one gets $\delta\zeta \simeq 300$ - 3000 K$\gg T$ and comparable to the Peierls energy gap of TaS_3 (1700 K). It is clear that such barriers have pronounced effect on the recombination rate. As E_T in thin samples is orders of magnitude higher its value in bulk ones, so thin samples are preferable for observation of photoconduction.

Another interesting feature of I-V curves of illuminated samples is a decrease of conduction with growth of the voltage in the creep regime which is clearly seen in Fig. 3 at $E \lesssim E_T$. From a formal point of view, this behavior corresponds to a negative contribution of creeping CDW to the total conduction of the illuminated sample. We believe, however, that this behavior results from the decrease of the concentration of photoexcited current carriers in the creeping regime, and possible contribution caused by CDW configuration variation due to change of current carrier concentration (configurational photoconduction). Such a decrease corresponds to an increase of the recombination rate of photoexcited current carriers due to modification and a time evolution of the potential relief in the creeping regime. Note that Ogawa *et al.* [7] attributed the growth of the total conduction at $E \lesssim E_T$ to the increase of CDW creep rate, i.e. to the opposite effect.

The illumination-induced variation of current carriers concentration must affect all static and dynamic properties of the CDW due to modification of the screening conditions. In particular, one can expect changes in CDW wave vector, relaxation rate of CDW metastable states, E_T, dielectric constant ($\epsilon \propto 1/E_T$ [1]), CDW transport coefficients, *etc.* Our results allows to verify for the first time the relations between the screening carrier concentration, CDW elasticity and E_T. As the transverse sizes of the our samples are smaller than the transverse CDW phase-correlation length, CDW pinning is one-dimensional [13]. In this case $E_T \approx (n_i w/K_\parallel)^{1/3}$, where n_i is the impurity concentration, w is the pinning potential, and $K_\parallel$ is the elastic modulus of the CDW [1, 13]. As $K_\parallel \propto 1/n$ [14], so $E_T \propto n^{1/3}$. Inset in Fig. 3 shows V_T^3 *vs* G dependence. The dependence is close to the linear one indeed.

It is well known that the energy dissipation for sliding CDW is provided by quasiparticles participating in screening of the time-dependent CDW deformations. Thus one could expect that growth of the current carrier concentration due to illumination would enhance the CDW conduction. However no noticeable modification of the CDW nonlinear conduction at $E \gg E_T$ is observed. Thus we conclude that the concentration of photoexcited current carriers diminishes with growth of CDW velocity. Noticeable reduction of the photoconduction starts already in the CDW creep region (Fig. 3).

The origin of the switching regime is a subject of wide discussions [15]. This regime is observed in $NbSe_3$ where the dielectrization of the electron spectrum accompanied by CDW formation is not complete and leads to the presence of free carriers in the system at low temperatures. Our results prove that the origin of the switching effect deals with the appearance of extra current carriers in the system despite their nature — natural ones as in $NbSe_3$ case or photoexcited ones as in TaS_3 case (present work) or in $K_{0.3}MoO_3$ case [7].

In conclusion, it was found that the light illumination of thin TaS_3 samples affects practically all their electrophysical properties. Namely, in illuminated samples we observed:

1) significant increase of the linear conduction;
2) strong nonmonotonic temperature dependence of the photoconduction;
3) highly nonlinear dependence of photoconduction on the light intensity;
4) decrease of conduction with growth of the voltage in the creep regime;
5) increase of the threshold field and suppression of the nonlinear conduction near V_T;
6) negligible effect on the nonlinear conduction at $V \gg V_T$;
7) appearance of the switching in I-V curves.
Observation of the photoresponse in the linear conduction is crucial for understanding the photoinduced changes in the CDW dynamics. Photoconduction in CDW conductors is much more complex phenomena than one in usual semiconductors because of very strong coupling between quasiparticles and the CDW. Possibility of photocontrol of quasiparticle conduction opens wide prospects for investigations of various static and dynamic properties of quasi-1D conductors.

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